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ASYNCHRONOUS OPTICAL SAMPLING FOR LASER-BASED
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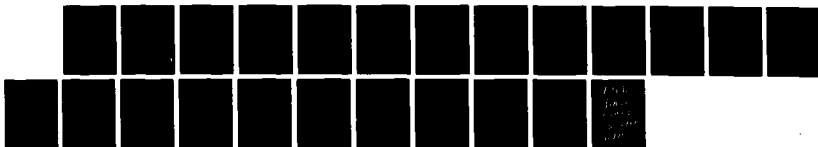
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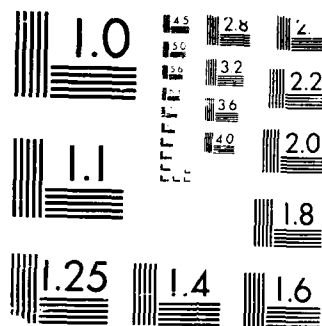
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REPORT DOCUMENTATION PAGE

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1a RESTRICTIVE MARKINGS None	
3 DISTRIBUTION/AVAILABILITY OF REPORT Distribution is unlimited; approved for public release	
4. PERFORMING ORGANIZATION REPORT NUMBER(S)	
5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR-TR- 88 - 0297	
6a. NAME OF PERFORMING ORGANIZATION Purdue University	6b. OFFICE SYMBOL (If applicable)
7a. NAME OF MONITORING ORGANIZATION Air Force Office of Scientific Research	
6c. ADDRESS (City, State, and ZIP Code) West Lafayette, Indiana 47907	
7b. ADDRESS (City, State, and ZIP Code) Building 410 Bolling AFB DC 20332-6448	
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR	8b. OFFICE SYMBOL (If applicable) NA
9 PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER AFOSR 85-0323	
10 SOURCE OF FUNDING NUMBERS	
PROGRAM ELEMENT NO 61102F	PROJECT NO 2308
TASK NO. A2	WORK UNIT ACCESSION NO
11 TITLE (Include Security Classification) Asynchronous Optical Sampling for Laser-Based Combustion Diagnostics in High Pressure Flames	
12 PERSONAL AUTHOR(S) G. B. King, N.M. Laurendeau, F.E. Lytle	
13a. TYPE OF REPORT Annual	13b. TIME COVERED FROM 12/15/86 to 12/14/87
14 DATE OF REPORT (Year, Month, Day) 1988, January 20	
15 PAGE COUNT 21	
16. SUPPLEMENTARY NOTATION	
17. COSATI CODES	
FIELD	GROUP
21	01
21	02
18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) combustion laser pump probe spectroscopy	
19. ABSTRACT (Continue on reverse if necessary and identify by block number) This report describes progress on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. The timing parameters for the current ASOPS instrument are described and consideration is given to the optimization of these parameters. The first ASOPS measurements in a combustion environment have been made on atomic sodium. These measurements are compared with laser-induced fluorescence measurements to demonstrate the viability of ASOPS as a combustion diagnostic.	
20 DISTRIBUTION/AVAILABILITY OF ABSTRACT <input type="checkbox"/> UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS RPT <input checked="" type="checkbox"/> DTIC USERS	
21 ABSTRACT SECURITY CLASSIFICATION Unclassified	
22a. NAME OF RESPONSIBLE INDIVIDUAL Julian M. Tishkoff	22b. TELEPHONE (Include Area Code) (202) 767-4105
22c. OFFICE SYMBOL AFOSR/NA	

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AFOSR-TR- 88 - 0297

ASYNCHRONOUS OPTICAL SAMPLING FOR LASER-BASED
COMBUSTION DIAGNOSTICS IN HIGH PRESSURE FLAMES

Progress Report
Air Force Office of Scientific Research
Grant No. AFOSR-84-0323
December 15, 1986 - December 14, 1987

Galen B. King*, Normand M. Laurendeau*, and Fred E. Lytle†
*School of Mechanical Engineering
†Department of Chemistry
Purdue University
West Lafayette, IN 47907



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Abstract

This report describes progress on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. The timing parameters for the current ASOPS instrument are described and consideration is given to the optimization of these parameters. The first ASOPS measurements in a combustion environment have been made on atomic sodium. These measurements are compared with laser-induced fluorescence measurements to demonstrate the viability of ASOPS as a combustion diagnostic.

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1. RESEARCH OBJECTIVES

The overall goal of this research is to develop and test a new combustion diagnostic for the quantitative measurement of both major and minor species in high pressure flames. The proposed technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond laser spectroscopy which will yield a better signal-to-noise ratio than laser fluorescence measurements in rapidly quenched combustion environments. Furthermore, ASOPS will allow determination of both quenching rates and state-to-state relaxation rates which are necessary for quantitative applications of both laser-induced and laser-saturated fluorescence at high pressure. The ASOPS technique produces a coherent signal-carrying beam and thus requires no more optical access to practical combustion devices than LDV measurements. By applying the proposed method, "real-time" concentrations of important species such as OH, NO and CO can be measured in flames at 1-20 atm.

2. RESEARCH STATUS

2.1 *ASOPS Methodology*

The specific goal of this project is to develop and prove the viability of the ASOPS technique as a combustion diagnostic in turbulent, high-pressure flames. ASOPS is a pump/probe method; however, it overcomes many of the temporal problems inherent to that method.^{1,2} Furthermore, corrections for the effects of quenching can be obtained on a time-scale necessary for practical combustion measurements.^{3,4} In the ASOPS method, rather than using a single Nd:YAG laser to derive both the pump and probe beams and an optical delay line to control the relative timing between the two pulses, two Nd:YAG lasers are used to separately drive the pump and probe lasers. Central to the method is the fact that the two Nd:YAG lasers are mode-locked at slightly different frequencies. The mode-locking frequencies of these two lasers are carefully controlled to maintain a constant beat frequency. In other words, a periodic relative phase walk-out exists between the pump and the probe

lasers. This has the same effect as varying the optical delay line in the conventional pump/probe method; however, the period of time needed to map the population of an excited energy level can be reduced from several minutes to less than a millisecond.

The process is illustrated in Fig. 1a, which shows the excited state population produced by several pump pulses and the temporal position of several probe pulses. Each successive probe pulse is delayed in time (relative to the pump pulse train) by a constantly increasing duration which is determined by the beat frequency of the system. Thus, each probe pulse samples the excited state population at a slightly later time than the preceding probe pulse. This is equivalent to varying the optical delay in a conventional instrument. The sampling process repeats itself when the cumulative delay equals the period of the pump laser. Hence, any modulation of the probe beam, resulting from the creation and subsequent decay of the excited state, repeats at the beat frequency of the system. Therefore, in contrast to a conventional pump/probe instrument, there is no need to modulate the amplitude of either beam to employ synchronous detection.

Fig. 1b illustrates the probe intensity which occurs upon stimulated emission from the excited state population shown in Fig. 1a. The net effect of the ASOPS technique is that a small amplitude waveform, which is directly related to the fluorescence decay of the species under study, is impressed onto the probe laser intensity. In essence, a temporal transformation of the excited state decay is performed with the time scaled by the factor $(f_{\text{pump}}/(f_{\text{pump}} - f_{\text{probe}}))$ where f is the repetition rate of the two lasers. The ASOPS technique is thus an optical analogue of the sampling oscilloscope.

Since the wavelengths of both the pump and the probe lasers can be independently controlled, specific electronic quenching rates between the excited and the ground states can also be determined. This assertion is valid because the stimulated emission (absorption) appears as a gain (loss) in the probe laser intensity; therefore, only those states which are directly connected by the probe laser wavelength are sampled. The loss/gain in the probe laser beam also provides information on the rate at which the depopulated lower level gains population from neighboring levels within the ground electronic state, or the rate at which the populated

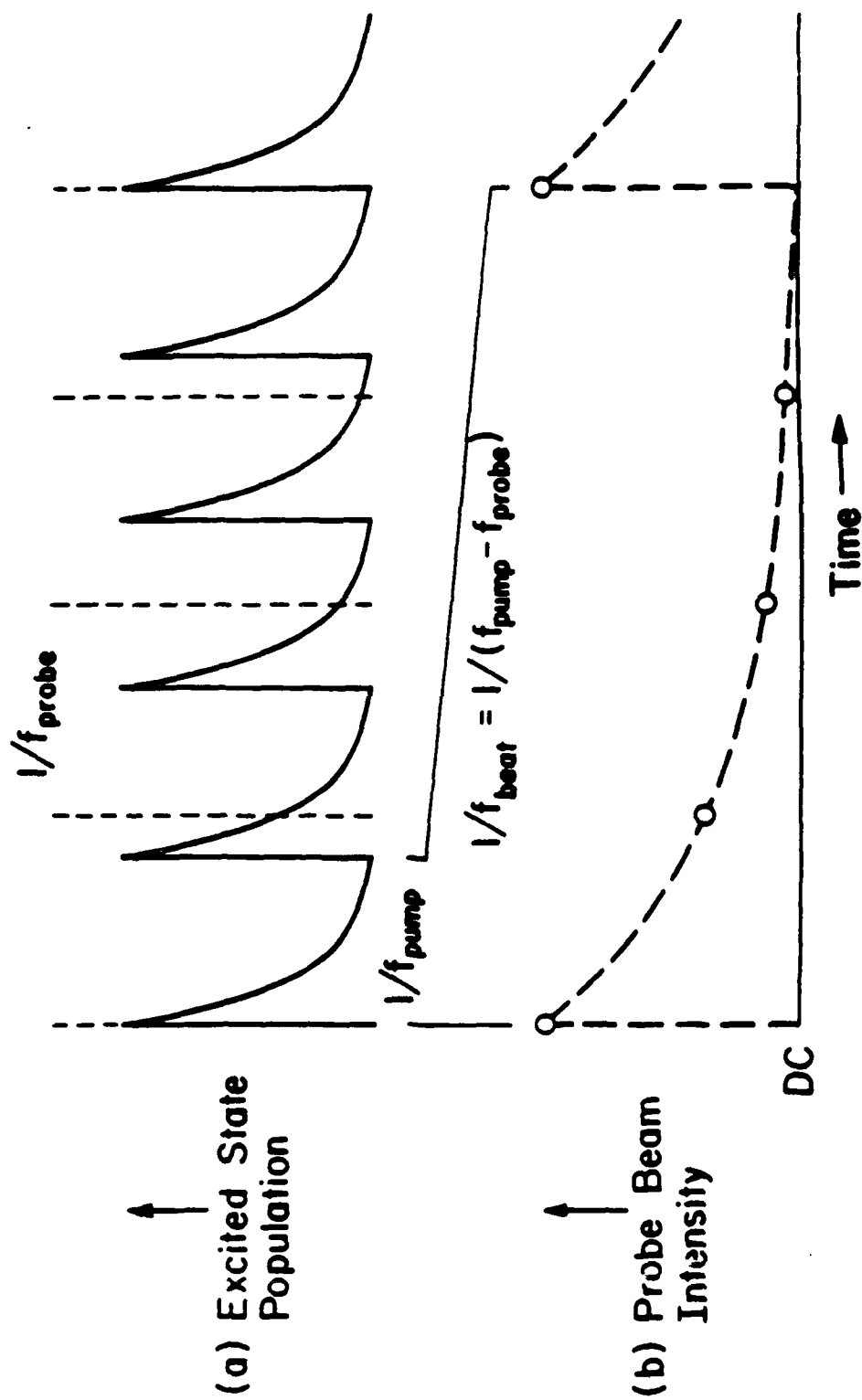


Figure 1. ASOPS timing diagram showing (a) excited-state population and (b) probe beam intensity. The probe pulses in (a) are indicated by the vertical dashed lines.

upper level transfers population to its neighboring levels within the excited electronic state. In other words, for molecular species the ASOPS technique can yield information about state-to-state relaxation rates within the upper and the lower rovibronic manifolds.

The above information is very valuable for laser-induced fluorescence (LIF) because absolute number densities are difficult to derive from fluorescence signals owing to poorly known collisional decay rates.⁵ Quenching rates can be calculated through knowledge of the rate constants for individual collisional partners and their respective number densities.^{6,7} However, the data necessary for these calculations are limited to only a few molecules and are typically obtained under different conditions of temperature, pressure and stoichiometry. Laser saturated fluorescence (LSF) can be used to avoid the quenching dependence^{8,9,10} but this technique is limited by the number of molecules that can be effectively saturated. Other investigators have used picosecond lasers for time-resolved LIF experiments on OH at atmospheric pressure, using either a streak camera^{11,12} or a time-correlated, single-photon counting scheme¹³ as the detection system. Although these experiments have the temporal resolution necessary to obtain quenching rates, the amount of time needed to obtain the data is much greater than the time-scale of turbulence. This is in part due to the limited repetition rate of the lasers which ranges from 1 Hz to 2 kHz. The slow triggering rates of the streak camera and the long times for photon counting also make these detection schemes undesirable.

The beat frequency of the ASOPS instrument is determined by the difference in laser repetition rates or $f_{\text{beat}} = f_{\text{pump}} - f_{\text{probe}}$. The inverse of the beat frequency, $1/f_{\text{beat}}$, represents the collection time for a single decay. The total number of points, N , sampled during the decay is determined by

$$N = \frac{f_{\text{probe}}}{f_{\text{beat}}}, \quad (1)$$

The difference in time between each sampled point within the decay profile is the sampling interval. The sampling interval and thus the temporal resolution is determined by the difference in laser periods, i.e.,

$$\text{Sampling Interval} = \frac{f_{\text{beat}}}{(f_{\text{probe}} f_{\text{pump}})}. \quad (2)$$

The period of the pump laser, $1/f_{\text{pump}}$, determines the free temporal range, which is the maximum time available for a single decay of the excited state population. Since the ASOPS signal follows a simple exponential, the duty cycle of the measurement is determined by the excited state lifetime of the species being monitored and the free temporal range. If we assume that the signal is zero after 3τ , where τ is the excited state lifetime, then

$$\text{Duty Cycle}(\%) = 100(3\tau f_{\text{pump}}). \quad (3)$$

The optimum beat frequency is influenced by a variety of factors such as collection time, samples per decay and sampling interval. Table 1 gives the above operating parameters for various beat frequencies.

Table 1. OPERATING PARAMETERS FOR VARIOUS BEAT FREQUENCIES			
f_{pump}	~82 MHz	~82 MHz	~82 MHz
f_{probe}	~82 MHz	~82 MHz	~82 MHz
Beat Frequency (f_{beat})	1 kHz	10 kHz	100 kHz
Collection Time (Single Decay)	1 msec	100 μsec	10 μsec
Samples per Decay (N)	82,000	8,200	820
Sampling Interval	150 fsec	1.5 psec	15 psec
Free Temporal Range	12.2 nsec	12.2 nsec	12.2 nsec
Duty Cycle (for $\tau = 1$ nsec)	25%	25%	25%

As can be seen, an increase in the beat frequency reduces the collection time and the number of points per decay, but increases the sampling interval. To make measurements on the time-scale of turbulence, it is necessary to collect enough data for adequate signal averaging in

approximately 1 msec. Currently, we are using a beat frequency of 10 kHz; hence, only 10 decays can be obtained in 1 msec. However, the beat frequency can be increased by up to a factor of ten by replacing one of the mode-locking crystals. An important goal of this research is to determine the beat frequency with the optimum signal-to-noise ratio. It is interesting to note that the free temporal range and the duty cycle are not affected by any change in the beat frequency.

2.2 Progress to Date

In the past year, we have made progress with respect to both the development of the ASOPS instrument and application of the technique to a flame environment. We first describe the components and configuration for ASOPS, as employed for our initial measurements of atomic sodium in a premixed flame. By using laser-induced fluorescence (LIF), comparisons are then shown between both relative concentration profiles and saturation curves obtained via LIF and ASOPS. The excellent agreement suggests that ASOPS has strong potential as a combustion diagnostic in practical combustion systems.

2.2.1 Instrumental

Since the ASOPS technique requires that the pump and probe lasers operate at slightly different repetition rates, the instrument must be constructed from two independent mode-locked laser systems. Both the pump and probe beams are currently derived from Spectra-Physics model 375B dye lasers, which are synchronously-pumped by frequency-doubled, mode-locked Spectra-Physics series 3000 Nd:YAG lasers. The mode-locking frequencies are generated by two Programmed Test Sources (PTS) model 160 frequency synthesizers (accurate to 0.1 Hz) operated in a master-slave (i.e., phase-locked) configuration to minimize drift in the beat frequency of the system. Both the pump and probe beams consist of an ~82 MHz train of ~10 psec pulses, tunable (using Rhodamine 6G) from 560-640 nm. The beat frequency is 10 kHz. Three plate Lyot or birefringent filters placed in the dye laser cavities result in bandwidths of 120 GHz ($\Delta\lambda = 0.14$ nm) for both the pump and probe lasers. Throughout the center of the tuning range, average powers in excess of 250 mW are obtained using 800-900 mW of pumping power.

A block diagram of the basic ASOPS instrument is shown in Fig. 2. Each beam is split as it emerges from the laser. The weaker fraction is used to generate a trigger signal at the beat frequency of the system. This is done by separately monitoring each beam with fast photodiodes (EG&G FND-100) and electronically mixing the outputs of the photodiodes with a double-balanced mixer (Anzac MD-141) operating in a high-speed gating mode.¹⁴ The mixer output is sent to a Texas Instruments LM311P comparator to provide a stable 0 to +5 V trigger pulse.

As shown in Fig. 3, the remaining fractions of both the pump and probe beams pass through polarization rotator-beam splitter combinations which provide a convenient means of varying the power in either beam. In addition, the vertical component of the pump beam is combined with the horizontal component of the probe beam at the second polarization beam splitter. Unlike Fig. 2, the beams pass colinearly through a focusing lens, the flame, and a collimating lens. Finally, the beams are separated by a third polarization beam splitter, after which the probe beam is monitored by a single photodiode (EG&G SGD-100A). Great care is taken to ensure that the pump and probe beams are completely separated at the final beam splitter, so that only the probe beam reaches the photodiode.

The photodiode output is filtered with a 4LE50CD Texscan 50 MHz low pass filter to remove the individual 82 MHz pulses, and is then directed to the signal processing system. For the temporal studies, a Hewlett-Packard 54100A digitizing oscilloscope, triggered at the beat frequency of the system, is employed. Each point of the oscilloscope signal is the result of 2048 averages. For the nontemporal studies, which include the concentration profiles and saturation curves, a Stanford Research Systems model 510 lock-in amplifier is used to monitor a single Fourier frequency component of the ASOPS signal.

The colinear configuration described above was chosen to compensate for the relatively large amount of noise present in our initial flame studies. Because of this configuration, the ASOPS signal is averaged over a pathlength of approximately 1 cm. In future experiments, spatial resolution will be enhanced by crossing the pump and probe beams, as shown in Fig. 2.

Basic ASOPS Instrument

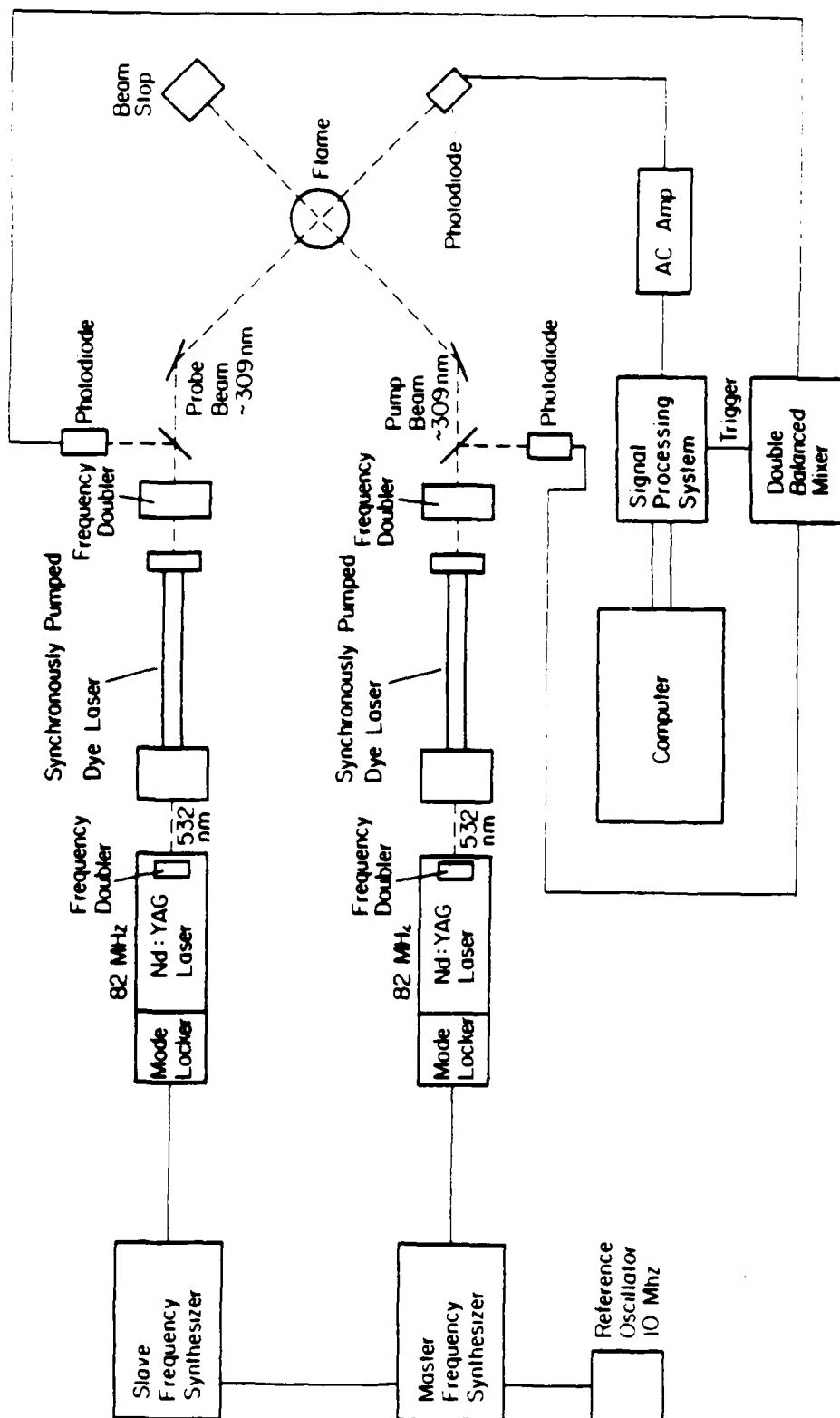


Figure 2. Block diagram of the basic ASOPS instrument.

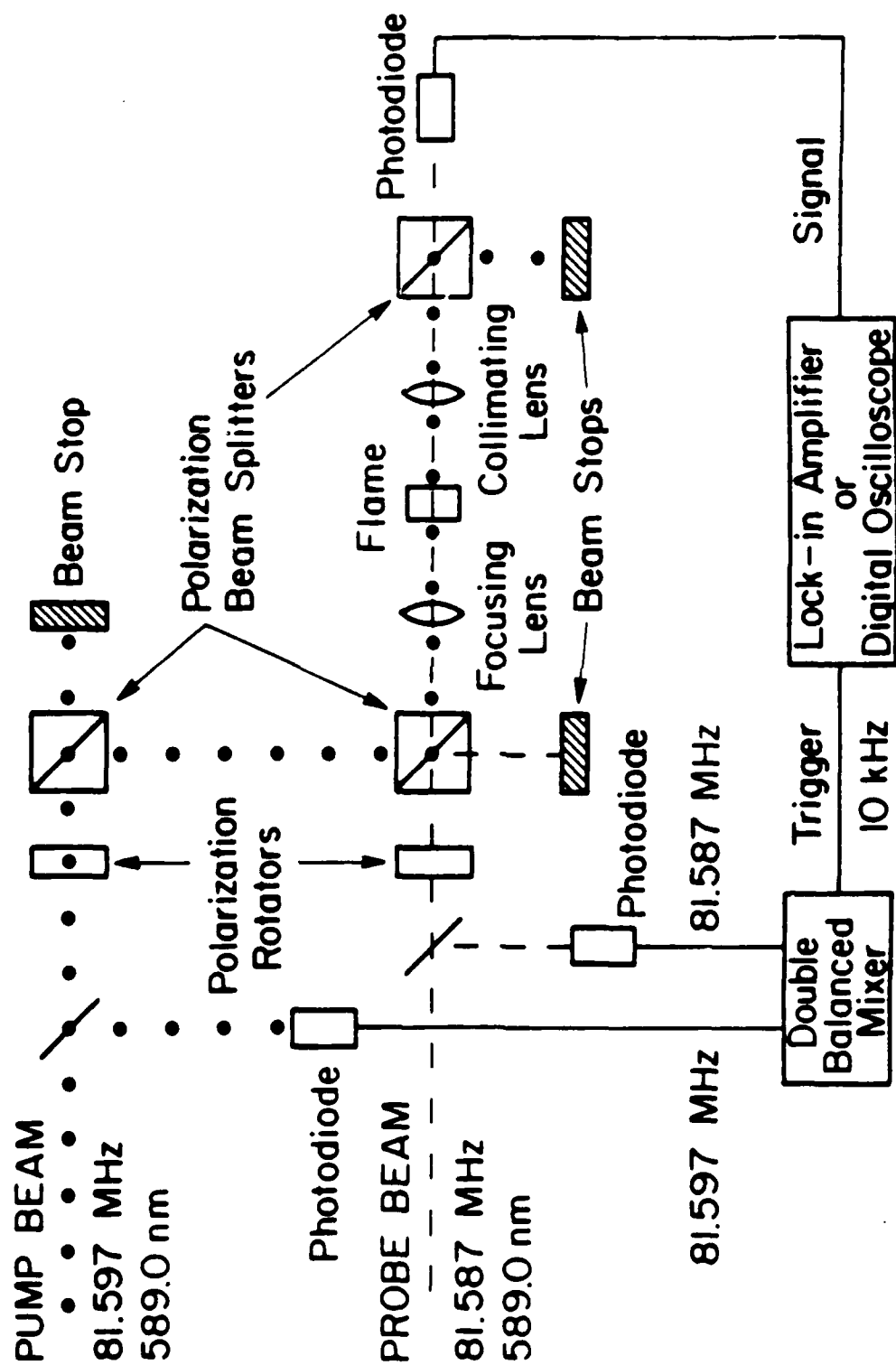


Figure 3. Block diagram of the ASOPS instrument used in the atomic sodium studies.

To provide a comparison with the ASOPS results, laser-induced fluorescence (LIF) was used to obtain additional concentration profiles and saturation curves. The output from one of the previously-described synchronously pumped dye lasers is amplitude modulated at 1 kHz using a PTi model OC 4000 chopper. The beam is then directed through a polarization rotator-beam splitter combination into the flame. Fluorescence is collected by a 15-cm focal length lens at right angles to the laser beam. To insure consistency, the fluorescence collection volume includes the same 1-cm pathlength through the flame used for the ASOPS measurements. A specially wired RCA 1P28B photomultiplier¹⁵ is used to detect the fluorescence. The photomultiplier signal is monitored using the lock-in amplifier at a reference frequency of 1 kHz.

An atomic absorption slot burner¹⁶ is employed to support an atmospheric premixed CH₄/O₂/N₂ flame into which a sodium chloride solution is nebulized. The burner provided fairly constant conditions for about thirty minutes, after which it became less reliable. Atomic sodium was chosen because of the large absorption cross section of its 3S_{1/2}→3P_{3/2} electronic transition. Since the 589.0 nm wavelength of the transition lies near the peak of Rhodamine 6G tuning curve, ample laser power was available to perform the saturation measurements.

2.2.2 Experimental

Figure 4 shows the ASOPS signal for atomic sodium; the signal compares favorably with that expected in Fig. 1. The parameters used in this study are summarized in Table 2.

Table 2. CURRENT OPERATING PARAMETERS FOR ASOPS	
f_{pump}	81.5970000 MHz
f_{probe}	81.5870000 MHz
f_{beat}	10 kHz
Pump Power	65 mW
Probe Power	10 mW
Amplification	100

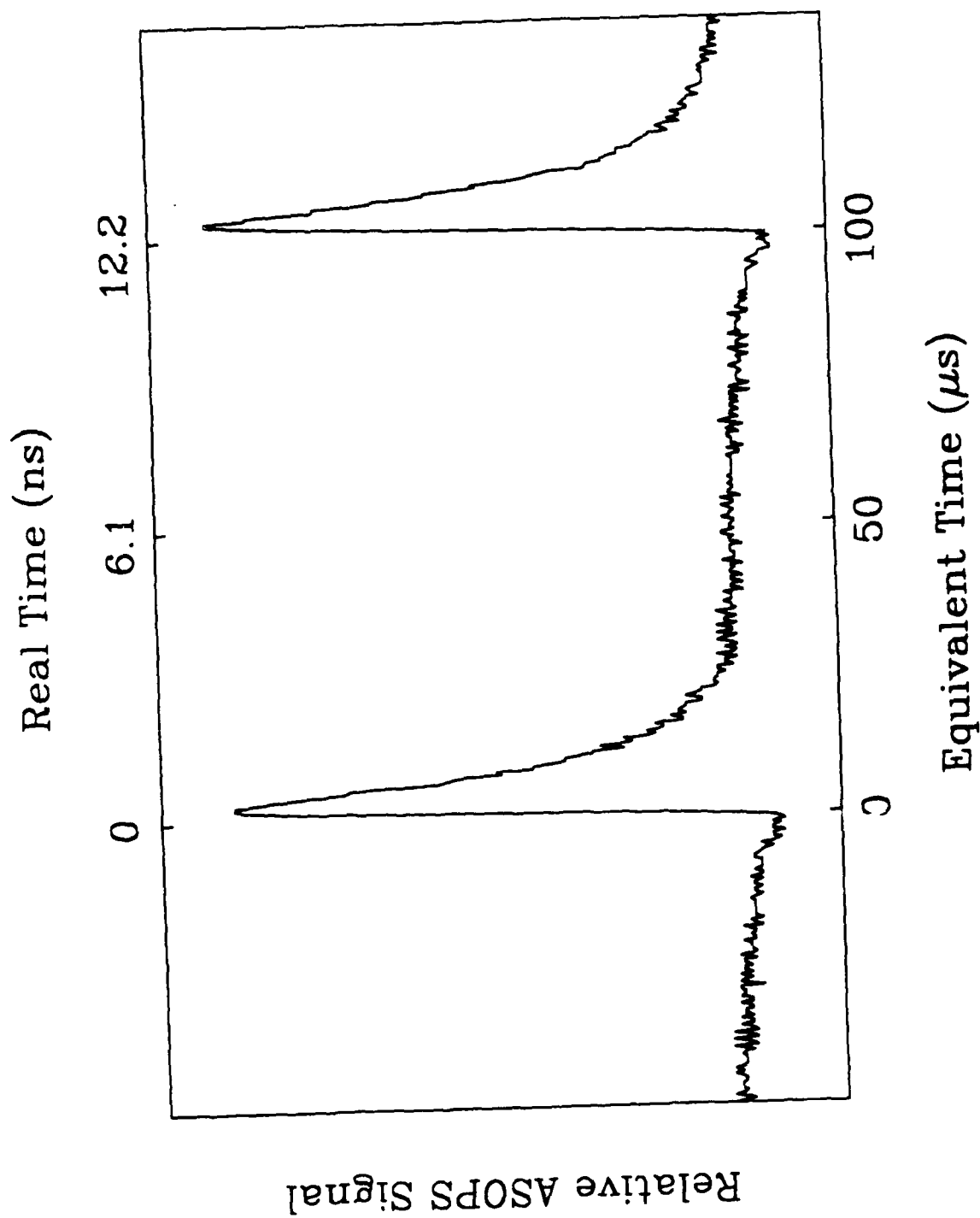


Figure 4. Temporal decay for atomic sodium obtained with both the pump and probe beams tuned to the D_2 transition (589.0 nm). The lower scale gives the equivalent time displayed on the oscilloscope, while the upper scale gives the actual time.

Based on a least squares fit, the equivalent decay time is 8.9 μ sec. Since the ASOPS technique performs a temporal transformation of the decay, the actual decay will be related to the equivalent decay by

$$t_{eq} = t \frac{f_{pump}}{(f_{pump} - f_{probe})}.$$

Thus the actual lifetime of the excited state under flame conditions is 1.1 nsec. This value is consistent with previous decay times (~ 1 nsec) reported in the literature.^{16,17}

Although the ASOPS signal in Fig. 4 appears to be an excitation curve, this is clearly not the case. Because the probe beam is directly connected to the ground state, it will be subjected to a combination of ground state absorption and stimulated emission, with the former dominating the latter. To understand the effect of stimulated absorption on the temporal profile of the probe beam intensity, consider the timing diagram in Fig. 5, where I_{pr}^0 denotes the intensity of the probe beam before the flame. The greater the ground state population (N_1) when the probe beam arrives, the greater the amount of absorption and the less the intensity of the probe beam after the flame. Hence, when the pump and probe pulses overlap, the ground state population is a minimum and the probe beam intensity is a maximum. This should be compared to the stimulated emission case shown in Fig. 1, where both the excited state population and the probe beam intensity are maximized upon temporal overlap of the pump and probe pulses. However, even though ground state absorption causes a decrease in probe beam intensity and stimulated emission causes an increase in probe beam intensity relative to I_{pr}^0 , the resulting temporal shape is identical in each case. Thus, it is clear that any combination of ground state absorption and stimulated emission will simply be an additive process with no distortion of the temporal decay.

The decay curve in Fig. 4 was obtained by averaging over 2048 separate single-decay curves. The peak signal-to-noise ratio of the average decay curve is 33. We believe that a considerable portion of this noise can be attributed to impedance mismatches at the trigger

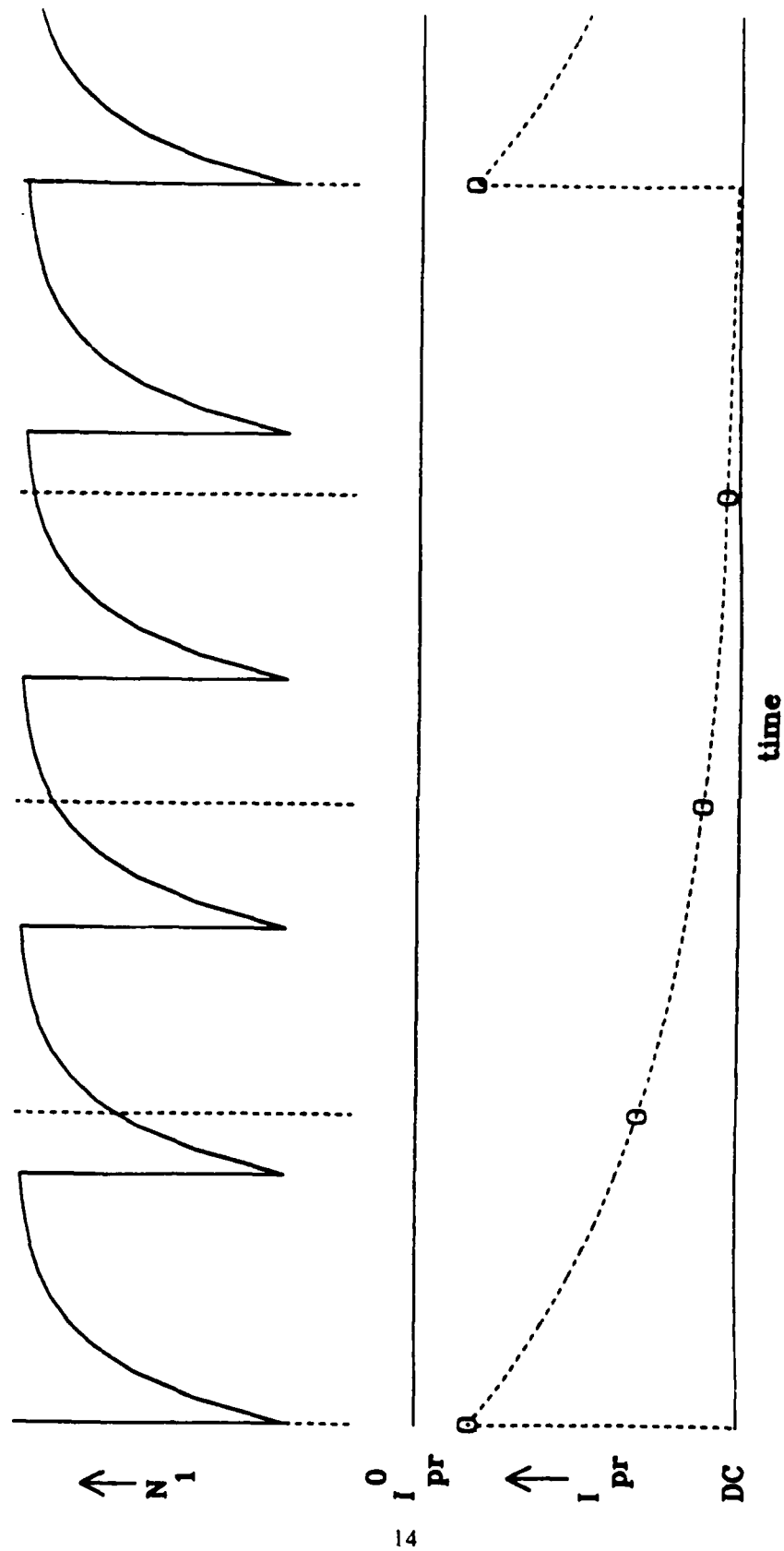


Figure 5. Timing diagram showing the effect of stimulated absorption on the ASOPS signal.

The DC level corresponds to maximum absorption of the probe beam.

photodiodes and at the double balanced mixer. Corrections are currently being made in these devices to improve the signal-to-noise ratio. Furthermore, suitable upgrades are possible with respect to the performance of both the Nd:YAG laser systems and the detection electronics.

The ASOPS signal in Fig. 4 contains an additional downward feature at the end of its free temporal range that is not predicted by Fig. 1b. We believe that this additional shape is due to competition between the pump and probe lasers. Figure 1a assumes that each probe pulse does not perturb the ground state before the arrival of the next probe pulse. However, this will not always be true in the present experiments because both the pump and probe lasers excite the same transition. For example, the 8199th pump pulse will arrive only 3 psec after the 8198th probe pulse. Thus for this pump pulse to see an unperturbed population, the disturbance caused by the previous probe pulse would have to disappear in 3 psec. This condition is obviously impossible to satisfy.

To further demonstrate the viability of ASOPS as a combustion diagnostic, both ASOPS and LIF were used to obtain both relative concentration profiles and saturation curves. The horizontal ASOPS and LIF profiles are plotted in Fig. 6. The pump and probe beam powers are 120 mW and 80 mW, respectively. The 120 mW pump beam is also utilized for the LIF measurements. As shown in Fig. 6, the relative concentration profiles obtained using each method are nearly identical. Furthermore, the signal-to-noise ratio for ASOPS was of the same order-of-magnitude as that for LIF. The ASOPS method yielded an inverse relative standard deviation of 34, while the LIF method yielded a value of 44. As was stated above, we expect the ASOPS performance to improve significantly with suitable corrections in the instrument.

The saturation curves obtained using both ASOPS and LIF are plotted in Fig. 7. For the ASOPS method, both the pump and probe beams are again tuned to the sodium D₂ transition. The pump beam power is varied while the probe beam power is held constant at 80 mW. For the LIF method, the probe beam is blocked, while the pump beam power is varied in a similar fashion. Once again, the ASOPS measurements correspond closely with the LIF measurements.

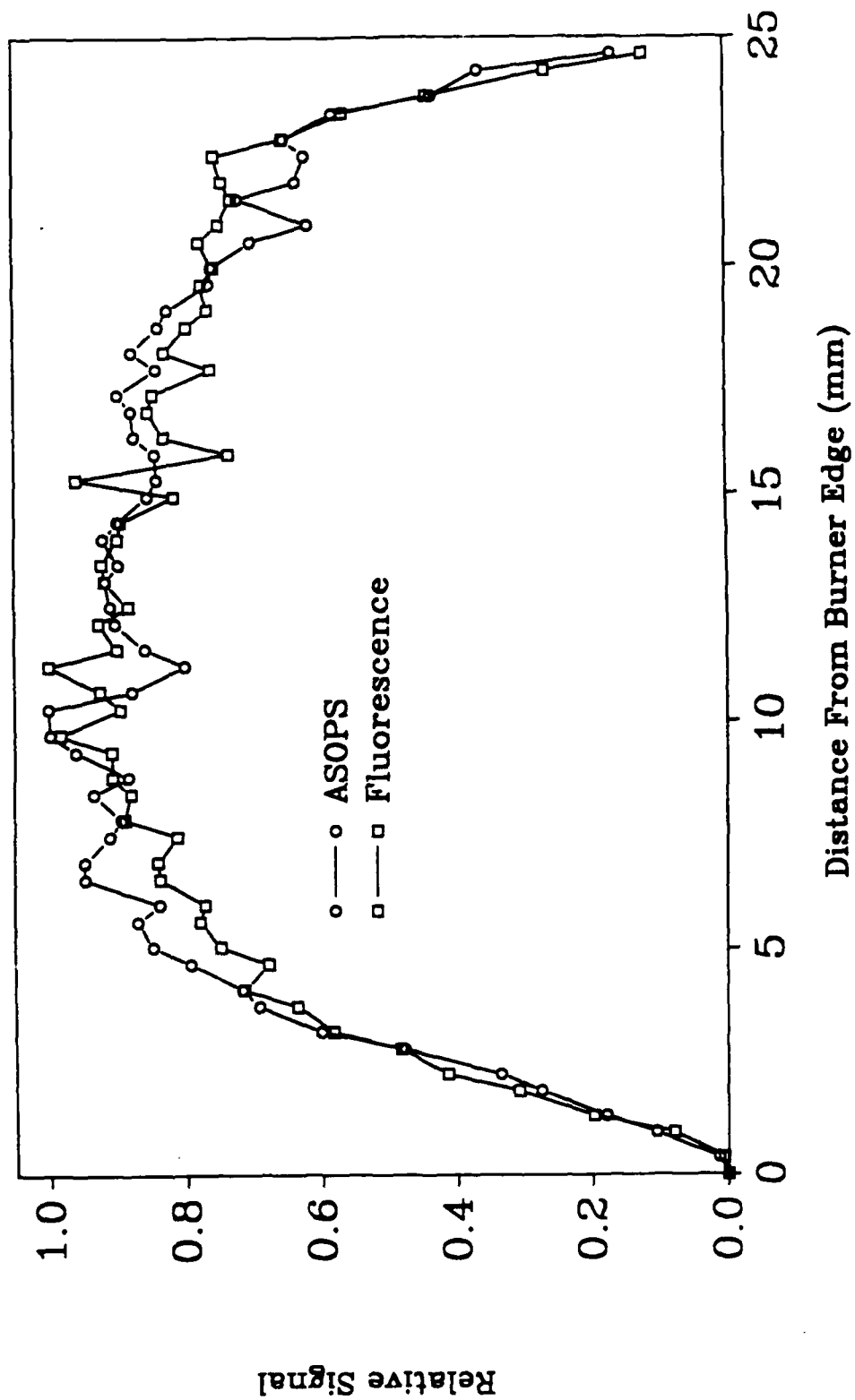


Figure 6. ASOPS and LIF relative horizontal concentration profiles. Measurements were taken ~0.6 cm above the burner.

SATURATION CURVE

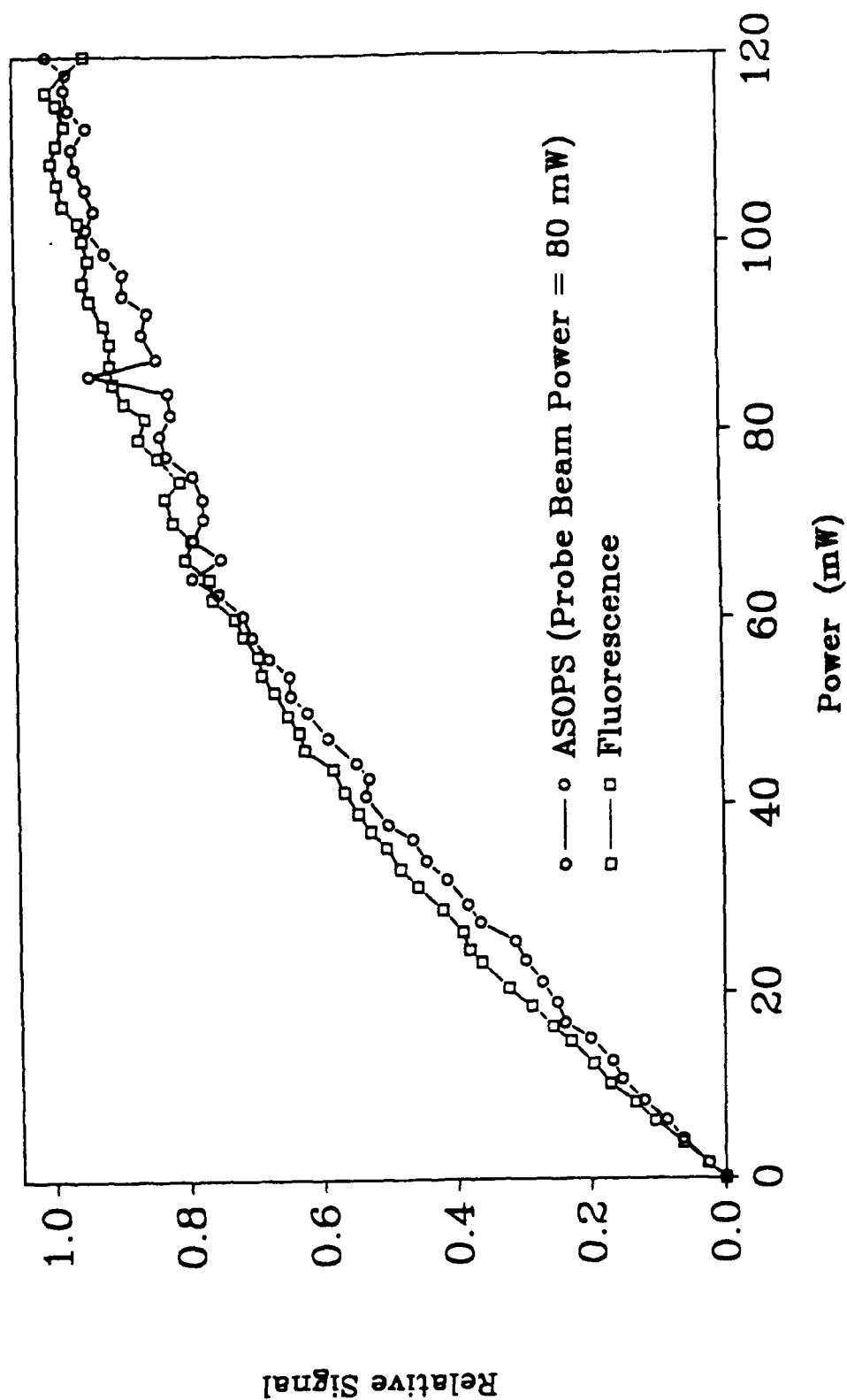


Figure 7. ASOPS and LIF relative pump-beam saturation curves. The ASOPS curve was obtained by varying the pump power, while the probe power was fixed at 80 mW.

2.3 *Current Research*

We expect to complete the sodium experiments within the next month. In order to extend the ASOPS technique to the UV portion of the spectrum, measurements will be performed in aluminum and/or gallium seeded flames. Both aluminum and gallium are well-described by a three-level model with two closely-coupled lower levels, in contrast to the two closely-coupled upper levels of the sodium atom. Furthermore, aluminum and gallium have strong resonant transitions in the UV. Hence experiments using these metals will provide an important intermediate step between atomic sodium measurements and future OH measurements.

A theoretical description of the ASOPS measurements is being developed using a rate equation approach. Both two- and three-level atomic models are being utilized in a computer program to generate expected ASOPS results.

Detailed analyses of the present instrument are being undertaken to categorize sources of noise. For example, a sizable impedance mismatch in the double balanced mixer is currently being rectified. A spectrum analyzer is being employed to detect the noise spectrum of each laser. This diagnostic tool allows us to modify the lasers to minimize noise originating in the lasers.

2.4 *Future Work*

Future work with the ASOPS instrument will involve frequency doubling both dye lasers to work in the UV. This will allow us to access the rovibronic transitions of the hydroxyl radical. Atmospheric premixed flames will be used to first determine absolute number densities and quenching rates for the hydroxyl radical. A new mode-locking crystal will then be employed to increase the beat frequency to 100 kHz, so that the data acquisition and averaging system can be optimized to take full advantage of the inherent speed of the ASOPS measurement.

3. PUBLICATIONS AND PRESENTATIONS

The following ASOPS papers have either been published or have been submitted for publication.

1. P. A. Elzinga, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, "Pump/Probe Spectroscopy by Asynchronous Optical Sampling", *Appl. Spectrosc.* **41**, 2 (1987).
2. P. A. Elzinga, R. J. Kneisler, F. E. Lytle, Y. Jiang, G. B. King and N. M. Laurendeau, "Pump/Probe Method for Fast Analysis of Visible Spectral Signatures Utilizing Asynchronous Optical Sampling", *Appl. Opt.* **26**, 4303 (1987).
3. R. J. Kneisler, F. E. Lytle, G. J. Fiechtner, Y. Jiang, G. B. King and N. M. Laurendeau, "Asynchronous Optical Sampling: A New Combustion Diagnostic for Potential Use in Turbulent, High-Pressure Flames", submitted to *Optics Letters*.
4. G. J. Fiechtner, Y. Jiang, G. B. King, N. M. Laurendeau, R. J. Kneisler and F. E. Lytle, "Determination of Relative Number Density and Decay Rate for Atomic Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling", submitted for presentation at the 22nd International Symposium on Combustion.

The progress of the ASOPS project has been presented at several meetings and seminars and has been well received by the scientific community:

1. "Pump/Probe Spectroscopy by Asynchronous Optical Sampling," American Chemical Society, Denver, CO. April 6, 1987.
2. "Asynchronous Optical Sampling: A New Generation of Picosecond Instrumentation," Pittsburgh Conference Lecture, Department of Chemistry, Pennsylvania State University, University Park, PA. April 22, 1987.
3. "Asynchronous Optical Sampling : A New Combustion Diagnostic for High-Pressure Flames," The Central States Section of the Combustion Institute, Argonne National Laboratories, Chicago, Ill. May 11-12, 1987.

4. "Asynchronous Optical Sampling," The Gordon Conference on Laser Diagnostics in Combustion, Plymouth, N.H. July 13-17, 1987.
5. "Two-Photon Spectroscopy of Molecules in Fluid Solution," Gordon Conference on Analytical Chemistry, New Hampton, N.H. August 10-14, 1987.
6. "Asynchronous Optical Sampling : A New Combustion Diagnostic Technique," Departmental Seminar, Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, N.J. September 29, 1987.
7. "Asynchronous Optical Sampling: A New Generation of Picosecond Instrumentation," Chemistry Department Colloquium, Purdue University, West Lafayette, IN. December 8, 1987.

4. RESEARCH PERSONNEL

Professors Galen B. King and Normand M. Laurendeau in the School of Mechanical Engineering and Professor Fred E. Lytle in the Department of Chemistry are co-principal investigators for this research. Prof. Yanan Jiang, a Visiting Scholar from the Department of Precision Instruments, Tsinghua University, Beijing, China, assisted on the project from January, 1986 to January 1988. Mr. Ronald Kneisler, a chemistry Ph.D. candidate, joined the group in January, 1986. Mr. Gregory Fiechtner and Mr. Yan Li, both mechanical engineering M.S. candidates, joined the group in July, 1986 and January, 1988, respectively.

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